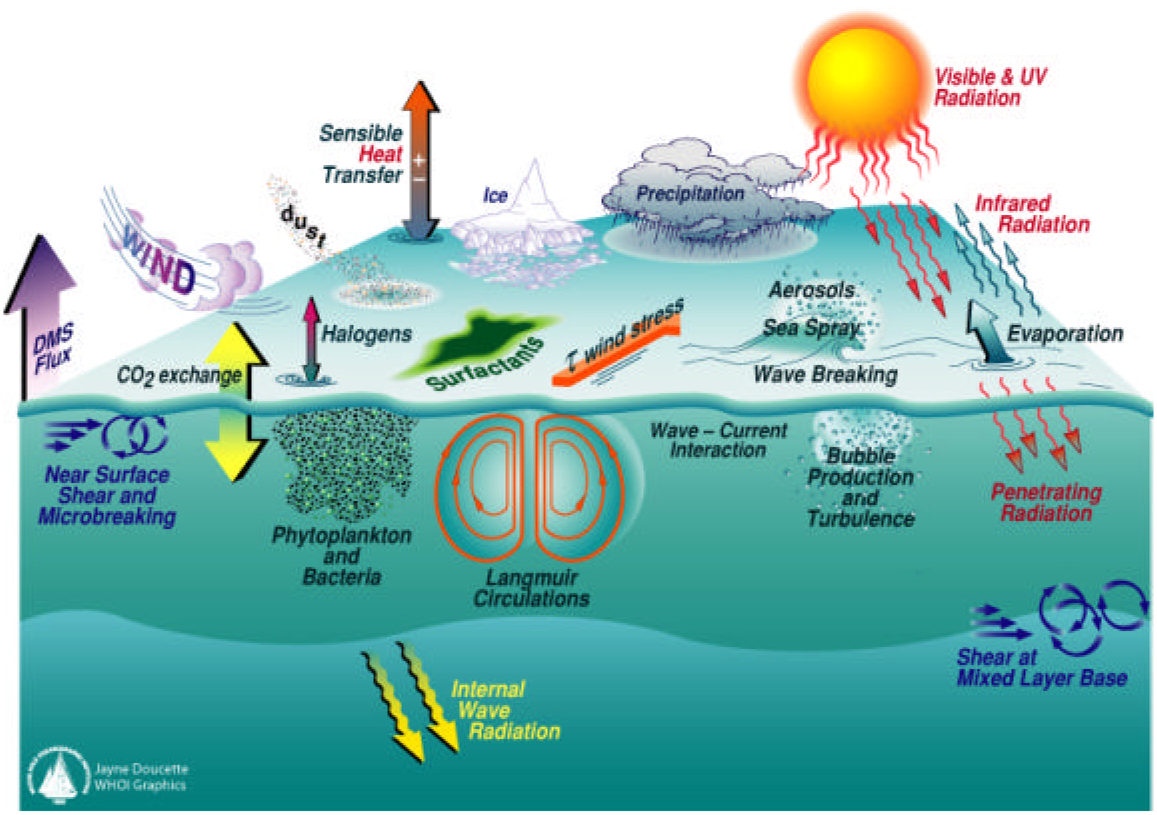
The United States Surface Ocean - Lower Atmosphere Study
(US SOLAS)

Workshop Report and Recommendations



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May 16-18, 2001
Bolger Center, Potomac, Maryland

Acknowledgments

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This report is available in electronic form at http://www.aoml.noaa.gov/ocd/solas

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Summary

This report summarizes the US SOLAS workshop deliberations held at the Bolger Center in Potomac MD, May 16-18, 2001. The goal of this open meeting was to lay the groundwork for the SOLAS program in the United States. The workshop consisted of plenary sessions highlighting the need for interdisciplinary study of the surface ocean-lower atmosphere realm for a variety of relevant biogeochemical and societal issues. Four working groups discussed science issues and priority projects that would benefit from a multi-disciplinary approach as advocated in SOLAS.

US SOLAS is a component of an international program endorsed by the IGBP and SCOR with the following as an overall goal:

> **To achieve a quantitative understanding of the key biogeochemical-physical interactions between the ocean and atmosphere, including how this coupled system affects and is affected by climate and environmental change.**

This international goal was adopted as a framework for the US effort. The working groups focused on those components that seemed of most interest to the US research community.

Each working group produced a report section that will form the basis of the US SOLAS science plan. The report describes the research issues and suggests future projects to further SOLAS science in the US. The suggested pilot projects exemplify the synergism between the topics.

The highlights and issues that came out of the working groups are as follows:

I. **Air-Sea Interaction and Transport Processes in the Atmospheric and Oceanic Boundary Layers**: Physical processes at the interface play a controlling role in transfer and transformations of Climate Relevant Compounds (CRCs). Quantifying fluxes of CRCs and the physical controls on transport are critical research areas where major advances may be achieved. Wet and dry deposition processes are poorly constrained, hampering the connection between atmospheric input and surface ocean biological responses. High priority issues include long-range research projects geared toward capturing episodic events, additional coastal region studies and technology development to improve flux measurements.

II. **Long-Lived Climate Relevant Compounds in the Surface Ocean-Lower Atmosphere Region**: These compounds have slow reaction rates in the lower atmosphere and the proposed research efforts are focused on constraining large-scale fluxes of CO_2, N_2O, and CH_4. Up-scaling process studies through opportunistic research cruises while fully integrating remote sensing capabilities are proposed. There is also a strong need to obtain basic physical and biological information on

transformation rates for many of the halogenated compounds that fall in the long-lived CRC category.

III. **Short Lived Climate Relevant Compounds in the Surface Ocean-Lower Atmosphere Region**: Tropospheric ozone, particulate matter and clouds are among the most powerful, but least understood elements of climate forcing. The SOLAS region is a major sink for ozone, and both a source and sink of particulate material containing organic matter, sulfur compounds and sea salt. Photochemical and multi-phase processes in both the upper ocean and lower atmosphere control the concentrations and distributions of these CRCs. The closely interlinked nature of their processing calls for a coordinated, multidisciplinary approach.

IV. **Atmospheric effects on marine biogeochemical processes**: Recent evidence that atmospheric deposition of trace constituents to the surface ocean can have a significant effect on upper ocean biological processes has fundamentally changed the understanding of biogeochemical cycling, nutrient limitations, and atmospheric versus sub-surface control. In particular, the strong interest in atmospheric deposition of iron and its possible effect on nitrogen fixation and enhanced biological productivity in the surface ocean as well as on the air-sea exchange of CRCs make this a high priority US SOLAS project.

An important goal of SOLAS is to provide observational, theoretical and modeling information for global-scale climate investigations with an emphasis on how air-sea exchange of heat, momentum and chemical species may influence atmospheric chemistry and climate. The air-sea exchange of CO_2 is but one example of the many different processes and feedbacks in the climate system that are intimately involved in surface ocean, lower atmosphere interactive dynamics. Facilitating this connection, several of the researchers involved in the development of SOLAS are also closely tied to the Community Climate System Model (CCSM) development effort, which is a multi-agency, multi-institutional effort to create a state-of- the-art Earth system model.

The US SOLAS effort will continue along two pathways. First, a steering committee will be formed to act as a clearinghouse for information on projects that focus on the lower marine atmosphere and surface ocean mixed layer. A particular focus will be the implementation of bilateral integrated studies of both the atmospheric and oceanic boundary layers. Second, the steering committee will interact with the community at large to advocate multi-agency endorsement and funding of dedicated projects. Since the efforts will be focused on multi-investigator process studies, we anticipate that the National Science Foundation will play a major, but not exclusive, role in supporting the effort. Within this context, the steering committee will foster interactions and cross-disciplinary announcements of opportunity between various funding organizations.

Recommended Scientific Objectives and Activities

The following sections summarize the science of the US SOLAS effort proposed by the meeting subgroups. More comprehensive reports from each group can be downloaded from http://www.aoml.noaa.gov/ocd/solas. The international science plan, which served as a guide for the US component, can be found at http://www.uea.ac.uk/env/solas. Several field studies are singled out as examples of high interest for US SOLAS. The final section of the report provides recommendations for steering group activities and logistics issues.

There is increasing evidence that the biogeochemical cycles for the building blocks of life, such as carbon, nitrogen, and sulfur, have been perturbed by mankind. These changes result in appreciable impacts and feedbacks in the surface ocean-lower atmosphere (SOLAS) region. The exact nature of the impacts and feedbacks are poorly constrained because of sparse observations, in particular relating to the interconnections between the major biogeochemical cycles and associated physical controls. It is in these areas that the interdisciplinary research approaches advocated in US SOLAS will provide the greatest impact on scientific understanding. US SOLAS research will focus heavily on the natural variability of key processes, anthropogenic perturbation of the processes, and the (positive and negative) feedbacks on biogeochemical cycles in the SOLAS region (e.g., Figure 1). A major objective is to integrate results from the process studies, large-scale observations, small and large-scale modeling and remote sensing efforts to improve our mechanistic understanding of biogeochemical and physical phenomena, and feedbacks.

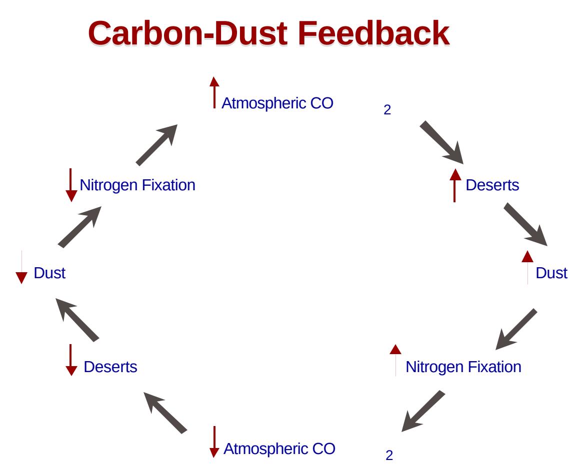

Figure 1. *The hypothesized link between atmospheric CO_2 levels and dust deposition is one example of the tight connectivity between lower atmosphere and surface ocean processes (courtesy of T. Michaels, USC).*

To test the SOLAS hypotheses and advance our knowledge of biology, chemistry, physics, and climate, experiment and theory must proceed simultaneously. Theories can be tested with numerical models, but models must be evaluated with measurements. *In situ* observations can provide specific properties, concentrations, and fluxes with known uncertainty, but only for limited areas and times. Our ability to make space-borne measurements of parameters such as wind speed, atmospheric ozone, aerosols, sea surface temperature, radiation, ocean color, sea surface height and sea state is improving. Recent advances in satellite-based remote sensing now also make it possible to measure atmospheric and oceanic properties such as atmospheric column concentrations of CO, CO_2, NO_2, H_2CO, BrO, H_2O, and aerosols. Instruments useful to studies in the SOLAS region include MOPITT, MODIS, MISR on TERRA, TESS and OMI on AURA, AIRS and SeaWiFS. European colleagues are developing complementary projects including GOME1 and radar altimeters on such as that on ERS2. Geostationary satellites may be particularly powerful for studying SOLAS processes, especially diurnal variations; these include NASA's GIFTS and the European GEOTROPE (GeoSCIA/GeoFIS) proposed to cover the Atlantic and Indian Oceans. These satellite measurements, especially when calibrated with "ground truth," can test the transport and spatial distributions of CRCs calculated in models. In this way, results from SOLAS studies will be expanded from specific measurements to general principles.

I. Air-Sea Interaction and Transport Processes in the Atmospheric and Oceanic Boundary Layers

Elucidating chemical and biological processes in the surface ocean and lower atmosphere requires a firm foundation in the understanding of physical processes that take place in this realm. Such processes are often parameterized with rudimentary physical forcing algorithms. Understanding the small-scale physical controls is critical to modeling these effects on larger scales. Processes of paramount importance to improving observations and modeling of the SOLAS region include:

- The physical processes that control air-water gas fluxes.
- Wet deposition by precipitation and dry deposition of particles onto the sea surface.
- Production of particulate material at the ocean surface.
- Mixing processes in the ocean mixed layer and atmospheric boundary layer and exchange with the stratified thermal interiors.

Significant advances in instrumental and modeling capabilities in the last decade make observation and verification of many of these processes tractable in laboratory investigations, Lagrangian field studies and modeling.

I-a. Physical processes controlling air-water gas fluxes

Gas transfer is regulated by molecular and turbulent processes in and near the molecular sublayers. For slightly soluble gases the aqueous sublayer controls gas transfer processes; for soluble or reactive gases the gaseous sublayer is of primary importance.

Small-scale processes such as short wind-waves, microscale breaking and near-surface turbulence are believed to control air-sea gas transfer. Any processes affecting small-scale turbulence, such as microlayer surfactants, or that short-circuit the diffusive sublayer, such as bubble entrainment, can have an effect on air-sea gas transfer of climate relevant gases. Significant advances have been made in probing the interface with optical and thermal methods deployed in field and laboratory environments. Advancements in direct-flux measurement techniques using accurate analyzers and micrometeorological approaches make it possible to determine fluxes concurrently with interfacial processes, and will greatly enhance understanding of the causal nature of the variability in gas transfer.

I-b. Wet and dry deposition onto the sea surface

Aside from gases, particles play an important role in the earth's radiation balance by transporting heat, momentum, and chemical and biological species in and out of the surface ocean. Two primary pathways of particulate material delivery to the surface ocean are direct deposition onto the sea surface (dry deposition) and scavenging, along with soluble gases, into water droplets and subsequent rainout (wet deposition). The mechanisms and effectiveness by which both processes occur is poorly constrained. Observations at process scale and modeling of the processes will constrain the deposition fluxes. This will improve our knowledge of atmospheric lifetimes of particulate constituents and related influences on multiphase chemical processes, direct and indirect climate forcing, and biogeochemical cycles in the surface ocean. Iron input through atmospheric dust deposition (Fig. 2) and subsequent biological response is a clear example of the importance of deposition processes on biogeochemical cycling. The process study information, together with data on aerosol optical depth and ocean color from satellite sensors such as MODIS, can constrain regional deposition effects (Fig. 3).

I-c. Production of particulate material at the ocean surface

Aerosols produced at the ocean surface play a critical role in the radiation balance and multiphase chemical processes that occur over the ocean. The mechanisms by which particles form at the interface are varied and poorly constrained. Reliable information on particle number, size, optical and chemical characteristics as a function of altitude is essential to properly model many processes in the SOLAS regime.

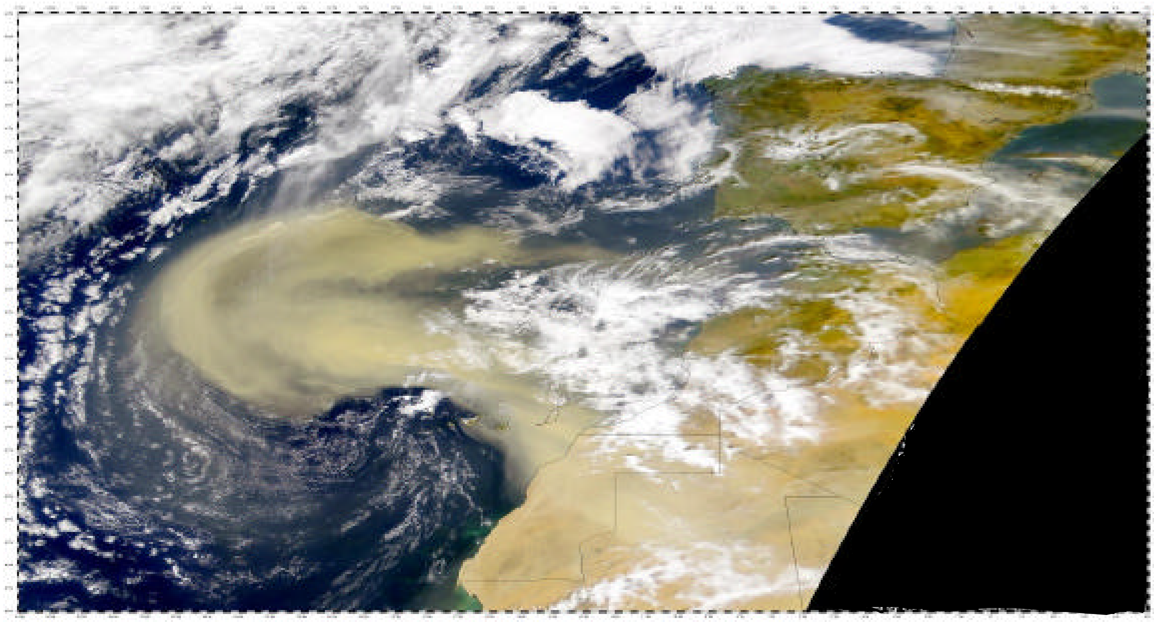

Figure 2. *A dust storm originating in the Western Sahara is shown in this true color image. Dust particles are carried westward by prevailing winds and can be observed thousands of kilometers from their origin. The impacts of the dust particles on surface ocean biology, heterogeneous (atmospheric) chemistry and radiation are poorly known. (Figure from http://www.visibleearth.nasa.gov).*

I-d. Mixed layer physics

Physical processes control, to a large extent, the supply of nutrients into the euphotic zone through dynamics occurring at the bottom of the ocean mixed layer. Internal waves, Langmuir cells and eddies all have a significant, but poorly quantified impact on thermocline – mixed layer interactions. Inputs of trace species often occur from the atmosphere or below the mixed layer. To understand feedback processes and forecast future changes in delivery and productivity, the specific origin of these species is of fundamental importance.

Similarly, dynamics at the top of the atmospheric boundary layer exert a fundamental control on boundary layer composition. Episodic exchanges between the marine boundary layer (MBL) and free troposphere are sources and sinks of reactive species to the boundary layer. Breaking waves and bursting bubbles can eject significant amounts of marine particulate material into the lower atmosphere. Transformations of gases and particles in the MBL lead to changes in cloud droplet size distributions and affect the Earth's albedo.

Studies aimed at improving the quantification of mixed layer dynamics range from utilizing probes to estimate turbulence and shear, to tracer studies in which the cumulative effect of the processes can be estimated. The key is to use methods and appropriate tracers that separate the physical processes from the chemical and biological transformations. Incorporating the basic physical information into models, which can then be used to predict the larger scale effects, is a powerful approach.

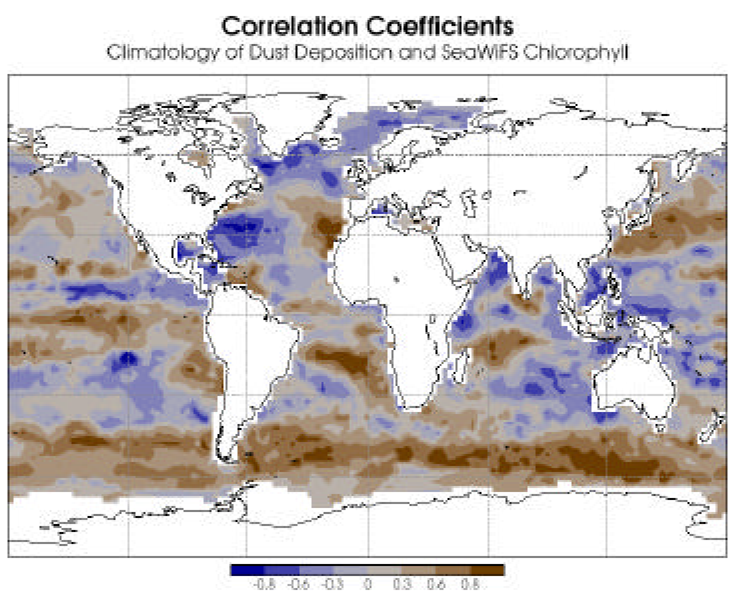

Figure 3. *The correlation between remotely sensed chlorophyll and modeled dust deposition is an example of innovative use of multiple approaches to assess the global effect of dust deposition on the open ocean and possible feedbacks as illustrated in Figure 1. The correlation suggests that iron input through dust might enhance biological productivity (courtesy of David Erickson III, ORNL).*

The greatest advances in boundary layer dynamics can be obtained by studies that cover a range of environmental forcing. This entails sustained observations of key variables from fixed platforms interspersed with more intense field campaigns to capture a wider range of dynamic responses.

II. Long-Lived Climate Relevant Compounds in the Surface Ocean-Lower Atmosphere Region

Fluxes of long-lived CRCs are strongly affected by boundary layer physics. Quantifying fluxes and inventories of these compounds relies largely on advances in boundary layer physics and modeling. One of the most important long-lived CRCs is CO_2, due to its climatic and societal relevance, and therefore CO_2 is a focal point in the SOLAS science plan. Comprehensive plans to study the oceanic and atmospheric CO_2 cycle are currently being developed as part of the NASA, NSF, and NOAA carbon cycle initiatives. Rather than duplicating these efforts, US SOLAS will augment plans under development to create an integrated multi-species program. Since the compounds in question, such as CO_2, CO, COS, N_2O, and many halocarbons, have relatively long atmospheric lifetimes, SOLAS will focus on air-sea fluxes, transformations in the surface mixed layer, and modeling.

US SOLAS can enhance the current efforts focusing exclusively on CO_2 by:

- Including the dynamics of radiatively important trace species in general circulation models to improve forecasting skills in determining the radiative balance.
- Obtaining basic physical and chemical characteristics of the compounds.
- Augmenting surface observations of the partial pressure of CO_2 with other CRCs.
- Providing a value-added product to surface CO_2 observations in near real-time by supplying key environmental information obtained from sources such as remote sensing and climatologies.

Numerical models of the climate system are rapidly moving toward full biogeochemical coupling. That is, the atmospheric concentrations of CO_2 and other CRCs are carried explicitly in the atmospheric flow, interacting with atmospheric radiation, and are directly influenced by surface source-sink processes such as air-sea exchange and material transfers with the terrestrial biosphere (e.g., Fig 4). This allows detailed feedbacks between biogeochemical and physical systems to influence climate prediction. For example, should any future shift in ocean circulation result in alterations to the air-sea exchange of trace gases (and aerosols), this would feed back into the system due to changes in the radiative balance of the atmosphere. Including other CRCs, such as N_2O, CH_4 and aerosols, in models of radiative forcing requires a better understanding of the basic biogeochemical cycles and air-sea exchange of these trace species.

To model the biogeochemical cycles of other long-lived CRCs, such as new anthropogenic halocarbons, requires consideration of the basic physical and chemical characteristics of these compounds. These include determination of solubility and diffusivity of gases as well as hydrolysis rates of the compounds in seawater. Such studies are best performed in controlled laboratory environments.

Several planning efforts focusing on CO_2 advocate monitoring programs on ships of opportunity including research vessels. This work should be augmented with other long- and short-lived CRCs, such as dimethylsulfide (DMS), halogenated and oxygenated organic compounds, whose oceanic sources and sinks are poorly constrained. Many of these compounds are produced or consumed by microbes but the specific functional groups involved in the transformations are unknown. Surface observations on research ships also offer the possibility for shipboard incubation studies to determine production and degradation rates of relevant CRCs for different biogeochemical provinces.

The larger observational and modeling community will benefit from incorporation of process studies into larger scale observations. This includes assimilation or merging of *in situ* data, remotely sensed products and climatologies. This is necessary to improve our understanding of the relationships and controlling processes. Much of the auxiliary information can be obtained from public sources, but the data have not been fully utilized. In part, the efforts involved in merging relevant remotely sensed or climatological data, such as wind speed, mixed layer depth, or sea surface height anomalies, into metadata sets are beyond the capabilities or time constraints of most individual research efforts. Providing support to the scientific community to use the products will facilitate interpretation of the data and aid in projects such as those interpolating measured quantities over larger space and time scales while providing large-scale process information on the factors that control, for example, surface water concentrations.

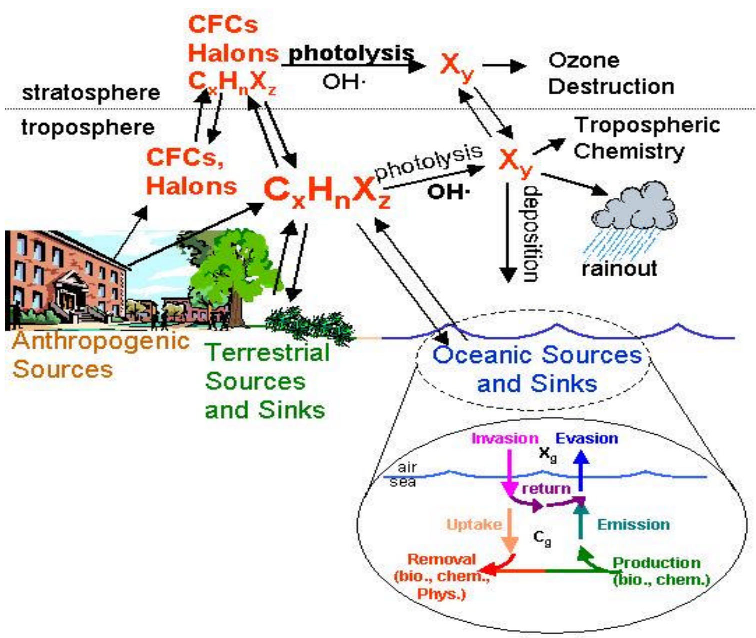

Figure 4. *Schematic of the major processes affecting boundary layer concentrations of halocarbons in the lower atmosphere and ocean mixed layer (courtesy of S. Yvon-Lewis, AOML/NOAA).*

III. Short Lived Climate Relevant Compounds in the Surface Ocean-Lower Atmosphere Region

The composition of the atmosphere also impacts climate change through varying fluxes of ozone and particulate matter – short-lived species whose concentrations change greatly in time and space. Ozone, a greenhouse gas itself, is involved in the biogeochemical cycles of many, perhaps most, CRCs. Aerosols contribute to climate forcing both directly as well as indirectly through a number of cloud-mediated processes.

Gases and particles with short atmospheric lifetimes (less than or equal to the residence time of atmospheric water, or about a week) generally show coherence on spatial scales smaller than 1000 km and reflect local or regional sources and sinks. However, the impact of these species on Earth systems such as climate can be global in scale. For example, the SOLAS region links transport of pollutants from one continent to another. Many short-lived atmospheric CRCs that originate from the surface ocean together with their reaction products play a major role in the chemistry and radiative properties of the lower atmosphere. Among these are sea-salt aerosols, halogen radicals (Cl, Br, and I), organic halogen compounds, NO, DMS, NH_3, condensed organic compounds and volatile organic compounds (VOCs).

Four broad but interrelated hypotheses emerged in the workshop as important to improving our understanding of the impact of SOLAS processes on climate and chemistry:

- Sea salt aerosols impact the cycling of trace gases and particles between the atmosphere and ocean.
- Marine sulfur and other elements appear to play a significant role in regulating Earth's climate.
- Fluxes of organic matter across the air-sea interface could play an important role in trace gas and aerosol cycling, photochemistry, and marine biogeochemistry.
- The photochemistry of the lower atmosphere and surface ocean is linked.

III-a. Impact of sea salt aerosols on the cycling of trace gases and particles between the atmosphere and ocean

Interactions among sea-salt aerosols, multiphase processes, and the fast photochemistry in the MBL are some of the most important climate-chemistry topics. However, the concentrations of sea-salt aerosol are not easily predicted. Winds play a role, but wind speed alone is inadequate to predict sea-salt particle concentrations. The physics of the surface ocean and concentrations of dissolved organic matter and surfactants may also be important.

The rate of ozone destruction in the remote MBL sometimes exceeds that attributable to ozone photolysis and standard RO_x chemistry, and bromine and/or chlorine atoms are implicated (see Fig. 5). The suspected mechanism involves acidification of sea-salt aerosols, formation and release of sparingly soluble Br_2, BrCl, and perhaps Cl_2. Recent studies suggest that iodine chemistry may likewise consume ozone. Iodine originates from photolysis of biogenic alkyl iodides, destroys ozone in a cycle involving marine aerosols, may be involved in new particle production, and accumulates as particulate iodate. The high rate of ozone removal observed during some field studies, if widespread, would constitute a major global sink. The relative loss of hydrocarbons has been used to infer a substantive role for Cl atom attack. If halogens play a major role in tropospheric O_3 and VOC chemistry, then they may also oxidize elemental mercury to make it more biologically active.

Nitric oxide, the limiting reactant in tropospheric ozone formation over most of the Earth, appears to be emitted by the ocean, although weakly as shown by field studies. If model simulations of heterogeneous oxidation of NO_x to nitrate are correct, then cycling of NO_x in the MBL is rapid and the marine source may be significant.

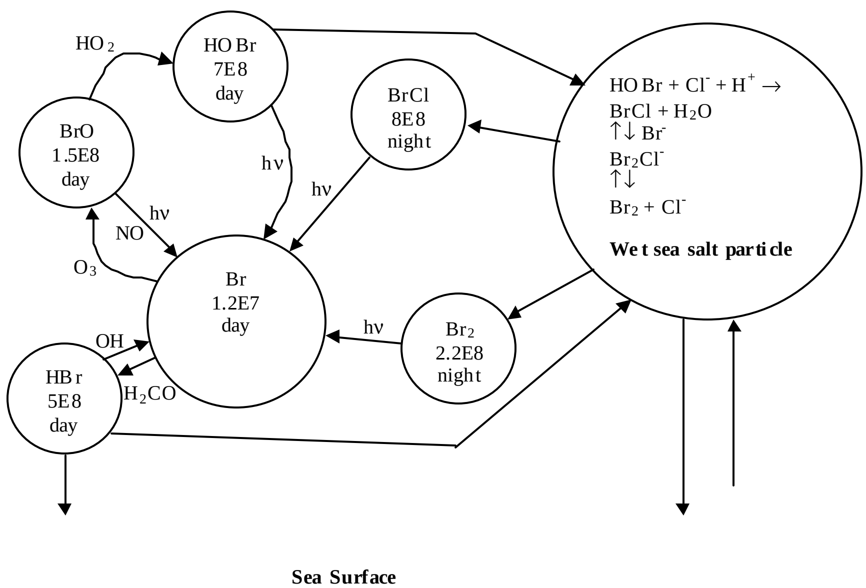

Figure 5. *Schematic diagram of multiphase Br chemistry in the MBL. The numbers in circles represent concentrations of the species indicated [molecules cm^{-3}]. The molecules on arrows show reactants and the large circle is a hydrated (wet) seasalt aerosol particle (courtesy of R. Dickerson, U. Maryland).*

Evidence for rapid halogen chemistry in the Arctic is particularly strong. Photochemical reactions on snow and ice may be a sink for O_3, but also a source of halogens, H_2CO, and NO_x. These processes may also impact the flux of trace species between the atmosphere and frozen surface and thus affect the reactivity of Hg and the long-term glacial record of atmospheric composition.

Detailed studies of HO_x (including OH) often indicate reasonable (factor of two or better) agreement between measurements and theory. Homogeneous gas-phase models cannot, however, account for the observed rate of ozone loss (as discussed above) or for concentrations of oxygenated organic compounds such as formaldehyde, H_2CO, acetaldehyde, methanol, and acetone. In the SOLAS region, these oxygenates are often found in much greater concentrations than non-methane hydrocarbons and should exert a significant influence on the oxidizing capacity of the atmosphere. Understanding the interactions of multiphase processes, the oxidizing capacity of the atmosphere and the photochemistry of the MBL and surface ocean require a deliberate, multidisciplinary approach.

III-b. The role of marine sulfur and related elements in regulating the Earth's climate

The direct and indirect influences of aerosols on Earth's radiative balance are currently the single largest source of uncertainty in anthropogenic climate forcing prediction. Particles enhance scattering and absorption of terrestrial and solar radiation. In addition to this direct effect, they can impact the thermodynamic stability of the atmosphere and the rates of photochemical processes in both the atmosphere and ocean. Anthropogenic aerosols further increase cloud brightness and connect to the hydrological cycle by inhibiting precipitation. In the SOLAS region there are three main classes of particles: sea salt, natural marine-derived sulfates, and aerosols transported from the continents. Particulate organic compounds from both marine and continental sources are associated with each class. The processes controlling the concentrations, radiative properties, and climate forcing of these aerosols are currently not well constrained.

Most DMS emitted from the surface ocean is oxidized to SO_2 and then, by both aqueous- and gas-phase pathways, a fraction of that is oxidized to H_2SO_4. Gaseous sulfuric acid condenses onto existing particles or can form (maybe in concert with marine NH_3) new aerosols that act as cloud condensation nucleii and strongly influence the size-number distribution of cloud droplets. This mechanism represents an impact of ocean emissions on precipitation and the radiative properties of the atmosphere. However, the processes controlling DMS emission are poorly understood. If cloud cover impacts the phytoplankton and the corresponding foodweb involved in DMS production, then a natural feedback cycle may be taking place.

The factors controlling the concentrations and fluxes of sulfur compounds in the SOLAS region remain uncertain. Major unanswered questions include the magnitude and distribution of significant atmospheric deposition of dust and nitrogen and the relative role of oceanic upwelling versus atmospheric input to the marine nutrient concentrations in the SOLAS marine region, as far as their effect on the biological communities involved in the cycling of sulfur compounds is concerned. These questions, also central to the boundary layer physics and biogeochemical foci, cut across disciplines. Several existing programs are actively investigating these processes from a marine perspective, and SOLAS-related initiatives will complement ongoing efforts while helping to provide an integrated framework for future experiments. Because of the complex web of interactions, a coordinated approach is needed with simultaneous measurements and models of atmospheric and oceanic biology, chemistry, and physics.

III-c. The role of fluxes of organic matter across the air-sea interface in trace gas and aerosol cycling, photochemistry, and marine biogeochemistry

Organic compounds are produced in large quantities by the oceanic biosphere, residing in the ocean as dissolved and particulate matter and in the atmosphere as VOCs and marine aerosols. The cycles of organic compounds are poorly understood, but may play major roles in controlling radiative properties, nutrient cycling, and cloud-aerosol interactions. The oceanic and atmospheric reservoirs are intrinsically coupled by air-sea and gas-particle exchange.

While the ocean is a source of both primary condensed organic material and VOCs, additional material is transported to the SOLAS region from continents. Both gas-phase and multiphase reactions are involved in chemical transformations and phase transitions. In seawater, organic matter helps control the aquatic light field and photochemical production of a variety of trace gases, nutrients, and reactive intermediates. Ocean opacity and color also play an important role in the Earth's radiative balance, and are coupled to climate change by non-linear processes that are difficult to predict. SOLAS objectives include studying the factors controlling the concentrations of organic constituents in the lower atmosphere and surface ocean and determining whether cycling through the atmosphere can impact organic matter sufficiently to change marine productivity. Key requirements to understanding the air-sea fluxes of organic matter include chemical and physical structure, thermodynamic properties, photochemistry, and distribution in the surface ocean. A priority is to develop new and improved measurement technologies for quantifying the concentrations and fluxes of VOCs.

III-d. Links between the photochemistry of the lower atmosphere and surface ocean

If the photochemistry of the lower atmosphere and surface ocean are intimately linked, then air-sea interactions may respond nonlinearly to climate changes. The trace gas and aerosol composition of the atmosphere controls (both directly and through changing cloud properties) the flux of solar UV and PAR reaching the ocean surface. Initial studies show strong surface ocean photochemical response to changes in UV-B radiation resulting from changes in column ozone content, indicating that other climate-related geophysical changes may also alter SOLAS region photochemistry. Available evidence suggests that deep convective clouds over the tropical ocean might transport organic compounds across the tropopause where they impact stratospheric ozone and aerosols. The organic composition of the upper ocean controls to a large extent the penetration of radiation and depth of the photic zone. Major unanswered questions include the spectra and quantum yields (number of product molecules formed per photon absorbed) of dissolved organic compounds, the impact of changes in ozone, aerosols and clouds on actinic radiation reaching the ocean surface, and the response of the surface ocean to changes in radiation.

IV. Atmospheric effects on marine biogeochemical processes

The need for an integrated atmospheric and oceanic program is particularly pertinent for the study of atmospheric impacts on marine biogeochemical processes. The hypothesis of aeolian dust deposition enhancing biological productivity, perhaps even in oligotrophic regions through a nitrogen fixation mechanism, is well documented. Several broader aspects are now being addressed under SOLAS and related planning efforts. Of fundamental importance are the following two questions:

- How is the ratio of phosphorous and nitrogen in surface water related to atmospheric fluxes of trace compounds?
- How do biogeochemical processes affect the air-sea exchange of CRCs and what are the possible feedbacks to radiative forcing?

IV-a. Atmospheric transport, processing and deposition of trace metals and the impact on marine ecosystems

Inorganic nutrients are found in the deep ocean in fixed stoichiometric ratios and many of the inferences on biogeochemical cycling are based on fixed "Redfield" ratios (with current widely used stoichiometry of P:N:C:O = 1:16:117:170). Organisms exhibit these ratios, but several biological functional groups/organisms have nutrient requirements that deviate significantly from the Redfield ratio. For instance, it appears that during episodic events, Redfield ratio constraints are often violated. Particles sinking from phytoplankton blooms often exhibit non-Redfield ratios and nitrogen fixing organisms in the open ocean that might be iron limited show enrichment in nitrogen with respect to phosphorous. Interestingly, although this increases the N:P ratio, the C:N ratio does not appear to change, making this pathway a possible natural CO_2 sequestration mechanism. Coastal regimes, in particular, are environments where there is riverine and aeolian input of fixed nitrogen and dust. Phytoplankton responses to these inputs often contain unique nutrient ratios. The issues of which (micro) nutrients control biological growth and the origin of the nutrients (aeolian, terrestrial, or marine) in different environments is a quintessential SOLAS question.

Just how important is the atmospheric transport pathway for these nutrients? In determining the possible implications of atmospheric input, there are a number of important issues that must be addressed. For example, it is not known what controls the temporal and spatial patterns of the atmospheric fluxes and what chemical processes control the bio-availability of the atmospherically supplied nutrients and bioactive elements that enter the surface ocean. This is particularly important for the nutrient iron, where the atmospheric input is related to the transport of iron-containing minerals from the continent, and where important chemical processes in the atmosphere during transport may affect the bio-availability of the iron in the ocean. Perhaps even more fundamentally, it is not understood how the nutrients derived from atmospheric input specifically affect marine biogeochemical processes and the relative availability of key limiting nutrients.

IV-b. The effect of biogeochemical processes on the air-sea exchange of CRCs and the possible feedbacks to radiative forcing

There is increasing evidence of biological processes exerting a fundamental influence on CRC concentrations in the SOLAS region. The biological functional groups responsible for production of many CRCs are not known. Even more fundamental is the issue of whether the CRCs are formed during production, respiration, or by bacterial processes. Biological processes can also directly affect the sea surface rheology through biological exudates that can concentrate at the sea surface. At high concentrations, these manifest themselves as surfactant layers. Biological production also affects the optical depth of the water and penetration of sensible heat. Therefore such processes must be included in models of surface forcing. In these cases, the biogeochemical cycles can act as feedbacks to the climate system. An optimal way of determining if such feedback mechanisms have operated in the past is by using paleo-proxies.

Perturbation and controlled-volume studies are particularly useful for investigating atmospheric impacts on the marine biogeochemical environment. The best-known perturbation studies are those of iron fertilization in HNLC (high nutrient, low chlorophyll) regions. Such studies to date have unequivocally shown an increased productivity following deliberate iron fertilization (Fig. 6) but they have not been of sufficient duration to determine the full response, particularly the export production of carbon. Moreover, other responses such as the possible reaction to dust deposition onto the oligotrophic gyres should be investigated. It is suggested that the warm oligotrophic ocean could have a productivity response through nitrogen fixation. Perturbation studies should not be limited to deliberate additions but should also include study of responses to natural perturbations, such as nutrient delivery from below the mixed layer during storms. Performing studies as a controlled volume will facilitate obtaining quantitative information.

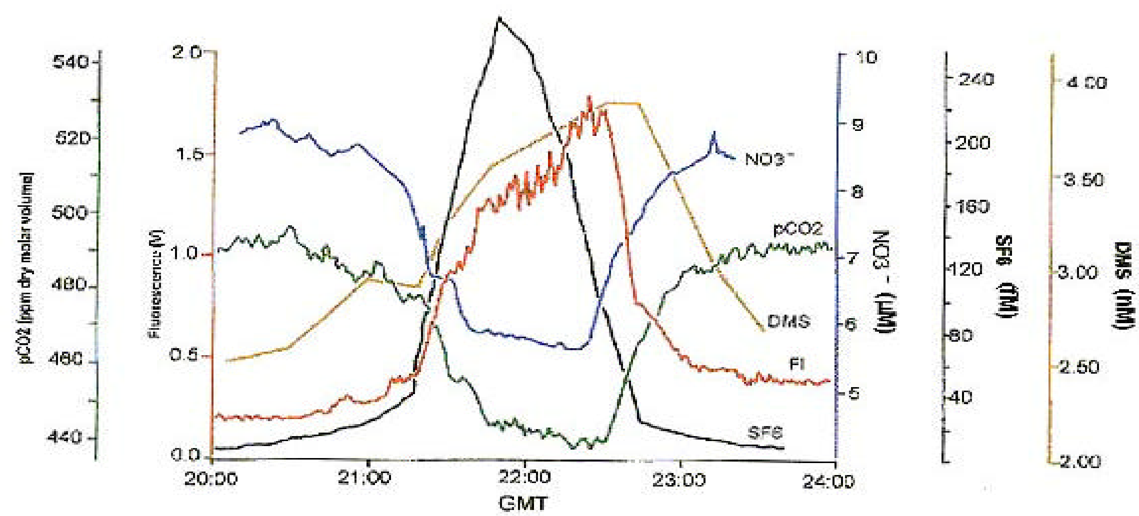

Figure 6. *Traverse across iron-enriched patch for a deliberate addition of iron and inert tracer SF_6 during IronEX II (from the SOLAS science plan, compilation of results).*

V. Suggested Studies

It is envisioned that US SOLAS will proceed on two parallel tracks. The US SOLAS community, through its steering committee, will advocate and coordinate select augmentation of established oceanographic and atmospheric research efforts with focus on connecting the two regions. However, full implementation of the SOLAS principles is best facilitated by planning projects from the ground up. Several of these conceptual "golden nuggets" are listed below. Others can be found in the working group reports. There are also some generic research issues ("enabling factors") that must be addressed to see the program off to a successful start. SOLAS activities will be greatly aided by the following enabling activities:

- Obtaining basic physical and biogeochemical information on a suite of CRCs in controlled environments.
- Including a wide suite of CRCs in models to improve their predictive skill as well as to provide critical information on the possible positive or negative feedbacks of the CRCs on the climate system.
- Infrastructure and field support for time series facilities in a variety of biogeochemical provinces. Such support is also advocated in many oceanic and atmospheric planning efforts. It is critical that there is a strong connection between both observational realms such that the forcing and feedbacks between upper ocean and lower atmosphere are clearly established.
- Technology development on measuring high frequency fluxes and concentrations of CRCs in the SOLAS region
- Setting up a coordinated data management system. Full utility of the suggested interdisciplinary programs will only occur if the data are readily accessible and properly quality controlled and documented. Comparisons of different studies which would be an integral part of the program will also be greatly aided by a unified data management system

V-a. Augmentation of Established or Planned Efforts

Several projects of direct relevance to the objectives of SOLAS have recently been undertaken or are scheduled for the near future. For future studies, augmentation with key measurements will greatly enhance their relevance as SOLAS-type projects and for studies that have recently been completed, support for multi-disciplinary interpretation is suggested. Examples of recently completed projects include the joint NSF/NOAA GasEx-2001 study, the multi-agency INDOEX study, the NSF/NOAA ACE-Asia study and the near concurrent PEM-West campaign funded by NASA. Synthesis projects that incorporate findings such as the largely ship based ACE-Asia study and aircraft-based PEM-West along with smaller studies such as the MBARI Western Flyer Monterey-Hawaii transect along with remote sensing could elucidate the oceanic response of large desert dust deposition. The data from the GasEx-2001 (Fig. 7) study along with collocated remote sensing data are well suited for higher order ground truthing and for upscaling results to estimate regional fluxes and factors controlling these fluxes. The three iron fertilization studies in the Southern Ocean should be interpreted in a holistic manner with focus on modeling and paleoclimatic interpretation of increased productivity events. Most of the projects mentioned included significant international components and collaborations.

Several field efforts are being planned that will take place prior to the full implementation of SOLAS. For example, a cruise under the auspices of the International Oceanic Commission (IOC) in the Western Pacific planned for 2002 focuses on the role of Asian dust deposition and the effect of trace metal delivery to upper ocean biota. Remote sensing efforts would be beneficial to obtain regional information. Establishing a quantitative link between atmospheric dust levels and upper ocean biological response studies of wet and dry deposition would be of merit.

On a larger scale, US SOLAS hopes to collaborate with the largely continental programs of the North American Carbon Program (NACP) and the Intercontinental Chemical Transport Experiment - A North American Mission (INTEX). Both studies aim to constrain fluxes of carbon dioxide and trace gases, respectively, on regional scales over the continental US. The oceans surrounding the continental US will be studied to deconvolve oceanic and terrestrial signals. In particular, residence times and transformation rates in the oceans differ dramatically from those in the atmosphere, and different cycles dominate each side of the interface. Severe knowledge gaps exist for photochemical oxidation of organic compounds in the surface ocean while the origin of high concentrations of oxygenated trace gases over the ocean remains a mystery. Multiphase processes such as oxidation involving sea salt aerosols, and even the processes maintaining the aerosol itself, stand as major unanswered questions in atmospheric chemistry. The interaction of continentally derived atmospheric species with marine aerosols also has the potential for major environmental consequences. Air quality programs that focus on coastal regions (e.g., Atmospheric Investigation, Regional Modeling and Prediction, AIRMAP) are addressing some aspects of this issue but not in a comprehensive fashion. Oceanic fluxes, in particular along the coast, must be constrained using *in situ* flux measurements and temporal and spatial extrapolation tools as provided by remote sensing. The coastal fluxes and atmospheric concentrations are a boundary condition for (transport) models that are an important tool for quantification and interpretation of the continental signal.

The significant (platform) resources available for NACP and INTEX can greatly aid US SOLAS projects with focus on the coastal US. US SOLAS aims to interact closely with the organizers of the projects to incorporate multi-disciplinary coastal boundary layer projects focused on CRCs.

A regional project that embodies the SOLAS objectives is the Sargasso Sea Ocean/Atmosphere Observatory (S2O2), which has formed to coordinate and enhance the contributions of the many marine biological, biogeochemical, hydrographic and atmospheric studies conducted in time series mode in and above the western North Atlantic. The S2O2 comprises several ongoing oceanic and atmospheric observation and modeling programs, which are in turn managed by several institutions. Primary objectives of S2O2 will be to maintain the vigor of the local time series programs and to provide a fluid and timely data stream to the international community of ocean and climate scientists.

Figure 7. *The NOAA Research Vessel Ronald H. Brown used for several studies of the SOLAS region including INDOEX and GasEx. Specialized infrastructure for these cruises such as a bow tower and booms sampled uncontaminated air and water.*

The US SOLAS steering committee will act as a source of information exchange to ensure that key measurements and interpretations connecting the lower atmosphere to the upper ocean are studied and performed while facilitating larger scale interpretation through optimal use of remote sensing and modeling, through national and international efforts whenever possible.

V-b. Golden Nugget Projects

Many of the SOLAS objectives can be facilitated by augmentation of planned or current efforts. However, to fully explore the interdisciplinary approach and foster interactions between the atmospheric and oceanic communities, dedicated projects should be implemented. The projects listed should at this point be considered as examples of the critical interconnectivity of atmospheric, interfacial, and upper ocean processes to study phenomena of critical importance in understanding the earth's system.

<u>Barbados Dust Deposition Experiment: BarDEx</u>

A project that covers many of the SOLAS objectives is one that was formulated several years ago to study the deposition mechanisms of African dust in the Western Atlantic and the upper ocean response. Deposition is not the sole requirement for an upper ocean biological response as, for instance, the bio-availability of trace nutrients is critical as well. Indeed, recent studies in the Pacific do not unequivocally show a biological response to dust deposition even in HNLC regions. A high priority within US SOLAS is to characterize wet and dry removal processes in regions where dust concentrations are known to be high. An experiment is proposed to take place in the tropical North Atlantic in the region of Barbados where trade winds carry high concentrations of dust during late Spring, Summer and early Fall. This experiment would have three overriding goals that encompass many SOLAS objectives:

- Characterize the removal rates of mineral dust and related aerosol particles via wet and dry deposition.
- Measure the deposition rate of dust to the ocean surface in individual rain events to follow the subsequent changes in the concentration of Fe and other species in the surface waters compared to dry deposition.
- Document biological and biogeochemical responses to these inputs.

The proposed project would include a longer duration, land-based phase for quantifying dust transport, mineral aerosol composition and size distribution, and (wet)-deposition fluxes along with shorter duration oceanographic process studies focusing on the deposition mechanism and the bio-availability of the trace constituents in the mineral aerosols. The effort would rely heavily on remote sensing products from sensors such as TOMS, MODIS, and SeaWiFS to characterize dust transport and upper ocean response. Established and new modeling activities would be entrained to focus on the effort. A synopsis of the study along with descriptions of related candidate projects can be found in Chapter 4 of the web based reports.

<u>Long term air-sea exchange studies</u>

Another example of a gold nugget project is long-term estimates of air-sea exchange of trace constituents over a broad range of environmental conditions for incorporation in regional and global scale models. A multi-year deployment at a fixed site designed to capture seasonal and interannual variability is recommended. This project would include intensive and continuous measurements of air-sea transfer of constituents with a range of solubilities, mass diffusivity, and controls on concentration in the atmosphere and ocean (e.g. chemical enhancement, surface reactive gases, catalytic reactions, and biological processes). To capture a large range of environmental forcing, including extreme events and biogeochemical responses, a mid- to high-latitude observational post would be desired. In addition to the central facility, arrays of moored and mobile instrument platforms would be deployed. Surface and near-surface mobile platforms can have conditional sampling routines built in to allow Lagrangian tracking of particular episodic events. The following studies are envisioned:

- Budget-based Lagrangian studies.
- Multiple tracer releases.
- Direct flux measurements.
- Ocean and atmosphere microlayer investigations.

Chapter 1 of the full report provides more information on such a project and other related projects of high relevance to US SOLAS.

Regional high latitude studies (Arctic)

A large Arctic regional program with strong SOLAS emphasis has been proposed on "Changing Environmental Controls on Coupled Chemical Exchange Between the Ocean, Ice, and Atmosphere in the Arctic."

The Arctic Ocean and its marginal seas play a fundamental role in the global ocean/climate system, due to deep water formation and its influence in thermohaline circulation, as well as its permanent sea-ice cover affecting Earth's albedo and associated radiation balance, and the distribution of freshwater. An important question is how the recently documented change in glacial and sea ice extent and thickness may impact the lower atmosphere and its composition, and how that may in turn feed back into changes in surface temperatures and Arctic biological processes.

Therefore simultaneous examination of processes that regulate the concentrations of gases in Arctic seawater, ice and snow leading to sea-air flux as well as ice- and snow-air flux is necessary. The broader scale impacts and ties to climate, transport and the fate of toxic pollutants will require a sophisticated approach to relevant Arctic-scale meteorology. The central role of a modeling component will be essential from the outset, the ultimate goal of which will be the refinement of an Arctic ocean-atmosphere sub-model (including physical transport as well as biogeochemical transformations) that can be included in comprehensive and predictive global climate-system models.

The following questions address some of the issues:

- Does halogen activation occur above/near open water/leads/polynyas, or is a frozen surface required? How would we go about testing this?
- How will changing sea-ice cover impact arctic boundary layer chemistry and the processing of pollutants, such as Hg?
- What is the role of the biota (phytoplankton; macrophytes; ice algae) in releasing photochemically active species such as organo-halogens and DMS, and how do we best pursue this question?
- How will climatic responses to changing sea-ice cover and the Arctic Oscillation impact the transport and processing of pollutants in/out of the Arctic?
- How should we treat the ice/snow surface so that chemistry on frozen surfaces can be treated quantitatively in models? What laboratory work is needed in this area?

VI. Interaction of US SOLAS with Other US Research Programs

There is significant synergy between the US SOLAS and other environmental programs developing in the US. The challenge is to optimize interaction and coordination both at international and national levels. US SOLAS will be closely aligned with the international SOLAS effort that has recently appointed a steering committee. Canada, Japan and Germany have already funded SOLAS efforts that will involve or offer opportunities for US participants.

In the US, continued interactions with the different agencies involved in the planning of global carbon cycle programs will occur. In particular, there is strong interest in developing interactions with the NASA carbon program, which focuses on remote sensing and coastal processes, and the Ocean Carbon Cycle Research group at NSF focusing on oceanic biogeochemical cycles of carbon and related parameters. Large-scale observational programs as planned in NOAA's sustained efforts include those with emphasis on the carbon cycle, trace gases and aerosols, and are good venues for hypothesis development and extrapolating findings to larger scales. SOLAS issues such as dust delivery, marine boundary layer transformations, and physics of the air-sea interface will be of direct benefit to the marine carbon oriented programs. The "carbon-based" projects envisioned in these programs can serve as a springboard for SOLAS efforts including the study of halogenated trace compounds, wet and dry deposition, and photochemical processes in the SOLAS region.

Aside from ocean-oriented programs, there are several continental and atmospheric efforts in which coordination with SOLAS would be beneficial. The NACP focus on constraining regional sources and sinks in North America will benefit from greater knowledge of oceanic CO_2 fluxes including regional interpolation mechanisms using remote sensing. Coastal towers and aircraft that include oceanic transects could be utilized for measurement of boundary layer dynamics and CRCs. The INTEX program, focusing on reactive trace compounds in the continental atmosphere, should provide many opportunities for both joint studies and data dissemination to expand the scale of observations from continental to oceanic and vice versa. As previously mentioned, the marine sources of many of the reactive halo- and hydrocarbons is either poorly constrained or completely unknown which causes significant uncertainty in the interpretation of large scale atmospheric chemistry models.

US SOLAS aims to work with the different programs under development. Several of the objectives brought forth in the international SOLAS plan have been adopted by other US planning efforts but often not in a full integrative fashion spanning both the ocean and the atmosphere. In such instances, US SOLAS aims to facilitate the connectivity between the oceanic and atmospheric reservoirs and research communities.

VII. Organizational Structure of US SOLAS

As manifested at the SOLAS community workshop, the concept of a surface ocean lower atmosphere study is well embraced in the oceanic and atmospheric research communities. In its current planning phase, US SOLAS is volunteer-based. The organizational aspects required to implement the effort as outlined requires continued participation and active scientific input from the research community.

It is envisioned that these efforts would be initially coordinated on a part-time basis by a senior scientific member with a 3- to 4-month per year commitment working in coordination with the international and US SOLAS Science Steering Committees. With input from agency representatives and consultation with the community at large, the SOLAS planning group will select a US SOLAS science steering committee to direct the implementation phase of the program. To assure continuity, some of the current planning group members will likely be part of the new scientific steering committee. Much of the committee work would be performed electronically and by teleconferences, with one meeting per year held in conjunction with a national meeting such as AGU and AMS. The request for funding to sustain the initial organizational component of US SOLAS efforts is estimated at the $200 K level. Funding would be sought through proposal submission to various agencies.

Appendix A: Narrative of the US SOLAS open science meeting

The meeting summary is abridged from the note in US JGOFS Newsletter (Vol 11, #2, June 2001).

Some 70 scientists representing a wide variety of oceanic and atmospheric disciplines attended the first community science workshop for the proposed U.S. Surface Ocean Lower Atmosphere Study (US SOLAS) at the Bolger Center in Potomac MD, May 16 - 18, 2001. The workshop attracted researchers in paleo oceanography, marine boundary-layer physics, ocean biogeochemistry, atmospheric chemistry and cloud physics. US SOLAS is a component of a larger international program, which was recently adopted as an interface program of the International Geosphere-Biosphere Programme (IGBP). The purposes of the workshop were to familiarize participants with the goals of SOLAS, to formulate relevant science questions for the U.S. component of the study and to discuss implementation strategies for the U.S. program.

The meeting started with presentations on international and national research programs that US SOLAS would be likely to interact with. International SOLAS is forming a scientific steering committee led by Peter Liss of the University of East Anglia, United Kingdom. Close ties are likely with the International Global Atmospheric Chemistry (IGAC) program, which has included studies of the marine atmospheric boundary layer as part of the Aerosol Characterization Experiment (ACE) and the Marine Aerosol and Gas Exchange Activity (MAGE).

Short presentations were made on the carbon-cycle science initiative programs of the U.S. National Science Foundation (NSF), the National Atmospheric and Oceanic Administration (NOAA) and the National Aeronautics and Space Administration (NASA). The research of quantifying the exchange of carbon dioxide (CO_2) across the air-sea interface is a major focus of these planning efforts as well as of SOLAS. Two continental field experiments, which include coastal regions, planned for North America in 2004, one focusing on atmospheric chemistry and one on the terrestrial carbon sink, offer possibilities for collaboration. A representative from Canadian SOLAS, the first national SOLAS program that has gotten underway, invited collaborations with studies planned in the sub-polar North Pacific and North Atlantic.

Four plenary talks offered context for the working groups, which convened to discuss science ideas and formulate priorities. Several overarching themes emerged from their efforts. For example, the need to quantify basic physical constants for a variety of compounds that have an important effect on climate. Issues include the solubility and diffusivities of halogenated trace gases; the characterization of organic compounds in the gas phase, in aerosol solutions, and in the surface ocean; biological consumption and production estimates for various compounds, and rates of photolysis.

Coastal margins, including the large arctic shelves, were singled out as requiring more attention. The high biological productivity, sedimentary processes, modified atmospheric chemistry of these regions contribute disproportionately to the fluxes of many compounds that affect climate, and critical interactions between continental runoff and the marine domain occur there.

The boundary-layer physics working group honed in on problems of quantifying the physical factors controlling fluxes and deposition of compounds, including gas-transfer velocity and wet and dry deposition mechanisms. Sustained coastal time-series measurements of fluxes of compounds that affect climate were suggested as one way to improve our understanding of the factors controlling gas transfer and deposition processes. The group also advocated studying the effects of high wind speeds on these processes.

The working group on the dynamics of long-lived compounds emphasized the need for regional flux information, particularly in the coastal regions. A lively discussion revolved around the question of US SOLAS participation in a survey of the partial pressure of CO_2 (pCO_2) in global surface water recommended in the large-scale oceanic and atmospheric carbon observing plan. The final recommendation was to support this effort and to strive for augmentation of pCO_2 surveys with near real-time flux estimates utilizing remote sensing and measurements of other compounds that affect climate. The group also noted a need for controlled studies of biological production and degradation rates of halogenated, nitrogen and sulfur bearing compounds.

The working group on dynamics of short-lived compounds had the daunting task of setting science goals with regard to a myriad of compounds that are reactive on both sides of the air-sea interface. Specific priority issues were identified and classified under four major, interrelated research foci: 1) Influences of multiphase processes involving sea-salt aerosols on the cycling of gases and condensed material between the atmosphere and ocean; 2) climatic effects of marine-derived S and other compounds; 3) the air-sea exchange of organic matter and related biogeochemical implications on both sides of the interface; and 4) linkages between photochemistry in the atmosphere and ocean.

The fourth working group focused on atmospheric effects on marine biogeochemical processes, including the cycling of iron and nitrogen. Highlighted issues were dust transport and deposition on the ocean and subsequent transformation of the iron in dust into bioavailable form. Participants noted that questions about wet and dry deposition and chemical transformation in the air phase were not fully addressed in other programs. They also discussed nitrogen fixation and deposition in different forms and noted the importance of coastal regions for studies of deposition. This group put forth two overarching themes for SOLAS research on upper-ocean biogeochemistry: How do atmospheric fluxes affect the nitrogen-to-phosphorus ratios of inorganic nutrients in the upper ocean? How do changes in biogeochemical processes in the surface ocean affect the air-sea exchange of compounds important to climate and thereby have a feedback effect on radiative forcing? These themes were broken down into more specific research objectives and field experiments that would address these issues.

All groups highlighted the importance of using information gathered by remote-sensing instrumentation to assess regional variability, to understand the effects of physical forces on biogeochemical processes and to extrapolate the results in time and space. The critical importance of maintaining and fostering long-term time-series observations was identified. Modeling priorities included coupling ocean biogeochemical process models with models of the atmospheric marine boundary layer for both long- and short-lived compounds. Participants urged the incorporation of both modeling and remote-sensing efforts into the initial stages of experimental design, site selection and the generation of hypotheses.

Appendix B: Participants at the US SOLAS meeting in Bolger, MD

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Appendix C: List of Acronyms and Abbreviations

AGU	American Geophysical Union
AMS	American Meteorological Society
CRCs	Climate relevant compounds
DMS	Dimethyl sulfide
HNLC	High nutrient, low chlorophyll regions
IGBP	International Geosphere-Biosphere Program
INTEX	Intercontinental Chemical Transport Experiment – A North American Mission
MBL	Marine boundary layer
NACP	North American Carbon Program
NASA	National Aeronautics and Space Administration
NOAA	National Oceanic and Atmospheric Administration
NSF	National Science Foundation
ONR	Office of Naval research
PAR	Photosynthetically available radiation
S2O2	Sargasso Sea Ocean/Atmosphere Observatory
SCOR	Scientific Committee on Oceanic Research
UV	Ultraviolet radiation
VOCs	Volatile organic carbon compounds
WCRP	World Climate Research Program